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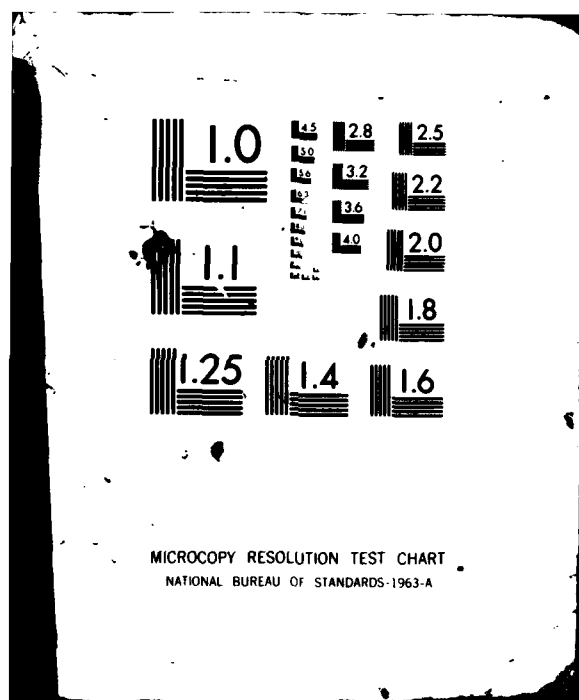   JUN 82   P HARRIS

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THE SHOCK FRONT RISE TIME IN WATER (U)

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INTRODUCTION

The question of shock front rise time is fundamental to the physics of dynamic high pressure and high strain rate effects in condensed media. The rise time question is currently THE basic question in shock wave physics. Applications range from a better understanding of the shock wave initiation of explosives to the design of radically new sensors for impact fuzing.

In a classic paper[1] Cowan and Hornig presented a study of the optical reflectivity of a shock front in a gas. The theory presented was for the linear case of small reflectivity and constant angle of incidence (within the shock front). The question of different shock structures (i.e., non-constant density gradients) was also considered. In a subsequent paper[2] Flook and Hornig attempted to measure the reflectivity of shock fronts in a number of transparent liquids including water. For the water experiments the reflectivity was too small to be measured.

This paper reports on successful shock front optical reflectivity theory and experiment for water at 5.8 kbar (0.58 GPa), and complimentary shock induced electrical polarization experiment and theory for the pressure range 20 kbar to 100 kbar. The LCWSL - Universite de Poitiers program has experiments performed in France (funded by the Commission Nationale Recherche Scientifique), while the ILIR program within the LCWSL funds the theoretical work.

The consistent interpretation of the reflectivity and shock polarization results is (a) a shock front rise time of approximately 10^{-12} sec throughout the pressure range of 0 to 100 kbar, (b) a transition in the source of the dominant polarization voltage to the alignment of unbonded dipole moments at approximately 35 kbar, and (c) in the unbonded region the average contributing dipole is perfectly aligned in the direction of shock propagation.

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SHOCK FRONT REFLECTIVITY

The shock front reflectivity theory and experiment have already been reported on elsewhere[3-5], and consequently that work will only be summarized here.

Figures 1 and 2 show both experimental and theoretical results for the reflectivity of a (laser) optical pulse from a propagating 5.8 kbar shock wave front in water. The experiments represent the first time that optically polarized beams (both parallel and perpendicular to the plane of incidence) were employed, and the theory, again for the first time, utilized an index of refraction and angle of incidence which varied within the thickness of the shock front.

The theory can be summarized via[4]

$$\text{Reflectivity} = \left| \frac{E_r}{E_o} \right|^2 , \tag{1a}$$

$$\left| \frac{E_r}{E_o} \right| = \sin\left\{ \frac{1}{2n_o} \int_{\infty}^{\infty} \frac{\partial n}{\partial z} (1 \mp \tan^2\theta) \cos\left[2\pi(\gamma_A - \gamma_B) \right] dz \right\}, \tag{1b}$$

$$(\gamma_A - \gamma_B) = \frac{2}{\lambda_n} \int_{-\infty}^{z} (n^2 - n_o^2 \sin^2\theta_o)^{\frac{1}{2}} dz, \tag{1c}$$

where the integration in Eq. (1c) is to a depth z within the shock front thickness. E is the electric field intensity, n and θ are local index of refraction and angle of incidence respectively, γ_A and γ_B are fractional optical phase angles, and the subscript "o" denotes preshock values while the subscript "r" denotes a reflected parameter. The - and + in Eq. (1b) denote parallel and perpendicular to the plane of incidence respectively.

Both constant gradient[3,4] (i.e. $\frac{\partial n}{\partial z}$ = const) and hyperbolic tangent[5] models for the index of refraction were used in Eqs. (1) with essentially the same analytic result. Aside from being the first accurate observation and analysis of shock front optical reflectivity for condensed media to appear in the literature, the results shown in Figs. 1 and 2 are important in that they provide a shock front thickness upper bound of approximately 400 A at 5.8 kbar.

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The 400 A upper bound is extremely important in that it pins the shock polarization results; to now we have only been successful in obtaining valid shock polarization data down to 20 kbar.

SHOCK POLARIZATION

The shock polarization results, including theoretical predictions at low pressures, are shown in Fig. 3. A modified "rigid four-point-charge" model[6] has been used in analyzing the data, and in making the low pressure predictions.

In homogeneous media computer molecular dynamics (CMD) calculations[6] each four-point-charge molecule interacts with neighboring molecules via an electrostatic potential (as well as via an oxygen-oxygen interatomic potential). In the derivation outlined below a single molecule is considered in the presence of a shock front induced torque field. The charges are used to calculate the molecular dipole moment. The model is illustrated in Fig. 4. Because only a single molecule is considered here (i.e. no neighboring molecules), inertial effects associated with the surrounding water structure is very crudely represented by placing hydrogen atom (proton) masses at those point charge positions. CMD or other detailed theoretical justification does not exist for such mass placement.

The fixed on the molecule primed coordinate system of Fig. 4 is imbedded in the laboratory (x, y, z) system where the +z direction is the direction of shock propagation (see Fig. 5). The calculation proceeds by using Euler angle transformations in order to find the torque about the line of nodes. The torque results in an equation of motion

$$\ddot{\theta} = \frac{2\ell}{3^{\frac{1}{2}} I} o f_n \; \sin\theta - \frac{k}{ma_{OH}^2} \int \theta \; , \tag{2}$$

where the $\sin \theta$ term is exact (if f_n is exact) and the second term is a crude restoring force with m being a proton mass, a_{OH} the oxygen-hydrogen intramolecular distance (1A), k is the hydrogen bond bending force constant, and I is the moment of inertia of the model molecule about the line of nodes with the oxygen atom as center.

The shock induced effective force (in the +z direction) on each hydrogen atom, f_n, used here is given by

$$f_n = \frac{\alpha \sigma}{n_o L_S} \; . \tag{3}$$

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σ is the peak stress amplitude, L_s the shock front thickness, n_o is the pre-shocked state number density of water molecules, and α is the fraction of the molecule force which acts on a hydrogen atom ($\alpha = 1/18$ for H_2O). Eqs. (2) and (3) represent a three dimensional version of a model first attempted by Horie[7].

Equation (2) has two interesting limiting solutions. One solution is where the shock induced torque is just balanced by the hydrogen bond bending restoring force so that $\ddot{\theta} = 0$. The second solution corresponds to the bond broken regime and thus has $k = 0$.

For $\delta\theta$ assumed small the $\ddot{\theta} = 0$ solution can be shown to lead to

$$<\cos\theta> = -\frac{\pi \ell_o m a_{OH}^2 f n}{(12)^{\frac{1}{2}} k I} \cdot \qquad (4)$$

Similarly the $k = 0$ solution can be put in the form

$$<\cos\theta> = -\frac{\pi \ell_o f_n t^2}{(48)^{\frac{1}{2}} I} \qquad (5)$$

In arriving at Eqs. (4) and (5) the angle average of $\cos\theta$ over a unit sphere has been used. Eqs. (4) and (5) give the average shock induced projection of the modified rigid four-point-charge along the direction of shock propagation.

Equations (4) and (5) are employed to arrive at the shock induced polarization P

$$P = n p_o <\cos\theta> \qquad (6)$$

where n is the number density of molecules which contribute to the observed shock polarization signals, and p_o is the four-point-charge molecule electric dipole moment ($p_o = 2.27 \times 10^{-18}$ esu cm from Fig. 4 with $|\vec{V}_1| = |\vec{V}_2| = 1A$ and $|\vec{V}_3| = |\vec{V}_4| = 0.8$ A). Because $L_s = U_s \tau_{sh}$ (with τ_{sh} the shock front rise time), Eqs. (5) and (6) predict P proportional to τ_{sh} for $t = \tau_{sh}$.

In turn P is used to calculate the prompt pulse current denisty[7,8]

$$J = \frac{K_o}{K} \frac{P (U_s - u_p)}{L_o} \qquad (7)$$

where K is dielectric constant, L_o is the sample thickness, and u_p the particle velocity. The form of Eq. (7) is appropriate to cgs units.

The t = 0+ voltage time derivative is given by[9]

$$\dot{V}(0+) = \frac{V(0+)}{\tau_{sh}} = \frac{P(U_s - u_p)}{\varepsilon_0} \tag{8}$$

where ε_0 is the vacuum permittivity (rationalized mks units).

POLARIZATION DATA ANALYSIS

@ Unbonded (i.e. k=0) molecules.

For σ = 100 kbar (10^{11} cgs), $t = \tau_{sh} = 10^{-12}$ sec, $n_0 = 3.47 \times 10^{22}$ cm^{-3}, $L_s = U_s\tau_{sh}$, $U_s = 5.05 \times 10^5$ cm/sec (from Ref. 10), and $I = 3.50 \times 10^{-40}$ gm cm^2

$$\langle\cos\theta\rangle = 0.82, \tag{9}$$

which is equivalent to saying that all of the contributing shock rotated molecules are almost perfectly aligned with the direction of shock propagation at $\tau_{sh} = 10^{-12}$ sec.

For $V(0+) = 0.2$ Volt one has agreement between the $\tau_{sh} = 10^{-12}$ sec voltage rise time and the right hand side of Eq. (8) providing that

$$\frac{n}{n_0} = 2.7 \times 10^{-3} \tag{10}$$

The Eq. (10) result is within a factor of 2 of the 100 kbar and corresponding Hugoniot temperature[10] ionic concentrations obtained by using the temperature dependent dissociation constants from Eigen and Maeyer[11] and the empirical conductivity versus hydrostatic pressure results of Holzappel[12].

The Eq. (10) result is also within a factor of 2 agreement with the 0.5% ion concentration at 130 kbar quoted by A.C. Mitchell el al[13] as a consequence of behind the shock front electrical conductivity measurements. It is also within a factor of 2 when compared with the ion concentrations obtained from behind the shock electrical conductivity measurements of Hamann and Linton[14] and an ion mobility of 3×10^{-3} cm^2 V^{-1} sec^{-1} (the atmospheric pressure 25°C value) from Eigen and Maeyer[11].

Applying $L_0 = 1$ mm, and $(U_s - u_p) = 3.06 \times 10^5$ cm/sec to Eq. (7) gives 0.2 Volt for the voltage predicted across a 50 Ω load by the experimental cell of 0.5 cm^2 cross section providing that $K=K_0$ and

$$\frac{n}{n_0} = 1.5 \times 10^{-3} \tag{11}$$

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are used. Although $K=K_0$ was used because these authors do not know better, it should be pointed out that in so far as it produces charges which can be separated in an electric field, a high degree of ionicity will increase the dielectric constant. An increased K would result in better agreement between Eqs. (10) and (11).

Using $\tau_{sh} = 10^{-12}$ sec and the above k=0 formulae yields the same very good agreement between theory and experiment down to 35 kbar. Near 35 kbar the slope of the polarization voltage versus pressure changes (see Fig. 3). The implication is a changing shock polarization mechanism.

(b) Hydrogen bonded molecules.

Applying the previously used numerical values at 10 kbar, and a k value[15] of 3.78×10^{-13} erg/radian^2, to Eq. (5) gives

$$\langle \cos\theta \rangle = -1.5 \times 10^{-4}. \tag{12}$$

In turn Eq. (12) yields, via Eq. (7) and the very reasonable bonded regime assumption that on the average each shock front enveloped water molecule contributes equally to the shock polarization current density (i.e. $n = n_0$), for the predicted prompt peak polarization voltage across a 50Ω load

$$V(50\Omega) = 15 \text{ m Volts}. \tag{13}$$

$K=K_0$ has been assumed.

For an extrapolation of the Fig. 3 experimental data to $\sim$ 6mV at 10 kbar, the left hand side of Eq. (8) becomes 6×10^9 Volts/sec while the right hand side is (from Eq. (12)) 8.7×10^9 Volts/sec. It is truly amazing that the very crude bending angle restoring force model utilized in Eq. (2) can give such excellent agreement between theory and experiment.

DISCUSSION

Employing the well known shock velocity values[10] and $\tau_{sh} = 10^{-12}$ sec one finds L_s (100 kbar) = 50A. That shock front thickness value is within a factor of 2 agreement with the computer molecular dynamics results[16] of Tsai and Trevino for shocks in dense monatomic liquids. 50A is consistent with the 5.8 kbar laser reflectivity value upper limit of 400A.

Because of the lack of experimental data in the low pressure region (i.e. <20 kbars), the 5.8 kbar reflectivity work becomes the glue which holds the work over the entire pressure range together. It can be shown that the 400A upper limit prevents the unbonded molecule mechanism from

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being the explanation for the observed low pressure polarization signals.

USSR researchers[17-20] have claimed a shock front related viscosity, η, value in the 100 kbar region of approximately 10^4 P. By using

$$\eta \sim L_s \rho U_s \qquad (14)$$

from Gilman[21], which should be valid in the 100 kbar range (ρ is mass density), one calculates $\eta \sim 0.2$ P at 100 kbar (from $L_s = 50A$).

There has been considerable discussion in the literature relating to the USSR value of 10^4P. While the reasons for the disagreement are not clear, it should be noted that Hamann and Linton[22], and Hamann[23], have consistently claimed that the USSR η values are incorrect.

The $\tau_{sh} = 10^{-12}$ sec and the corresponding claim with respect to molecular orientation within the shock front (100 kbar to 35 kbar range) represents a major advance in the interpretation of shock front physics. Second, the detail and accuracy of the experiment and theory for the shock front optical reflectivity serves as the low pressure anchor for the entire program reported on here. Further, it should be noted that the 400A upper limit reflectivity result limits the many USSR claims that such reflectivity measurements can predict molecular dimension shock front thicknesses; the thickness is too small in comparison to the optical wavelengths to yield sufficient theoretical accuracy for those claims.

That the bond broken regime dominates the polarization signals at high pressures is not surprising; Graham[24] has advanced the concept that shock induced bond scission is the agent responsible for shock polarization and increased electrical conductivity in polymeric solids. At the moment, however, it does not appear that bond breaking is the polarization producing agent in the 10 kbar region.

ACKNOWLEDGEMENTS

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REFLECTIVITY (PERCENT)

▲ DATA ⊥ POLARIZATION

● DATA ∥ POLARIZATION

——— THEORY

$(\Delta n)_f = 6.54 \times 10^{-2}$

$(\Delta n)_f = 5.94 \times 10^{-2}$

$(\Delta n)_f = 5.26 \times 10^{-2}$

10^{-1}

$\beta = 0.5$

$\beta = 0.2$

10^{-2}

A
B
C

$\beta = 10^{-2}$

$\beta = 10^{-1}$

G   F E D

GRAZING ANGLE (90° MINUS ANGLE OF INCIDENCE)

10^{-3} 0   10   20   30

DEGREES
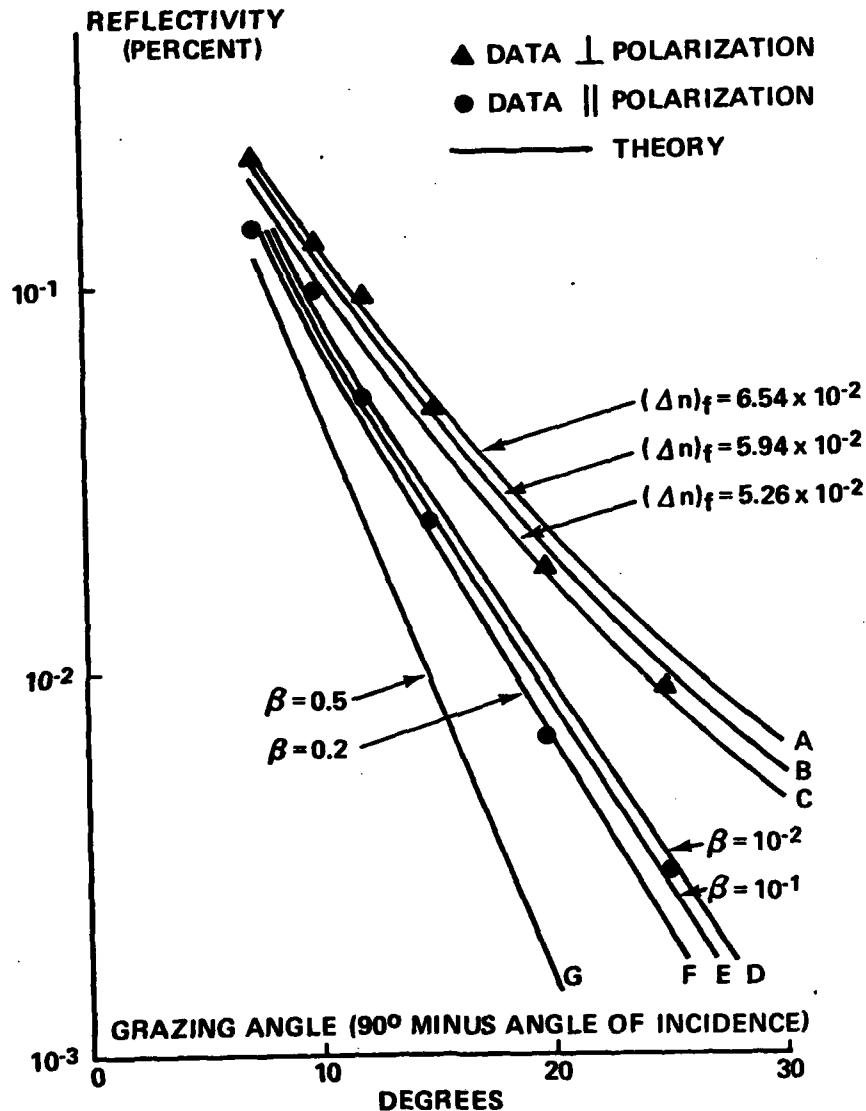
Figure 1. Theory and experimental data for shock front reflectivity at 5.8 kbar in water. Curves (A), (B), and (C) are for ⊥ polarization and $\beta = 0.1$. Curves (D) through (G) for ∥ polarization and $(\Delta n_f) = 5.26 \times 10^{-2}$. $\beta \equiv \frac{n_0 L_0}{\lambda_0}$, and $(\Delta n)_f$ is the total change in index of refraction across the shock front.
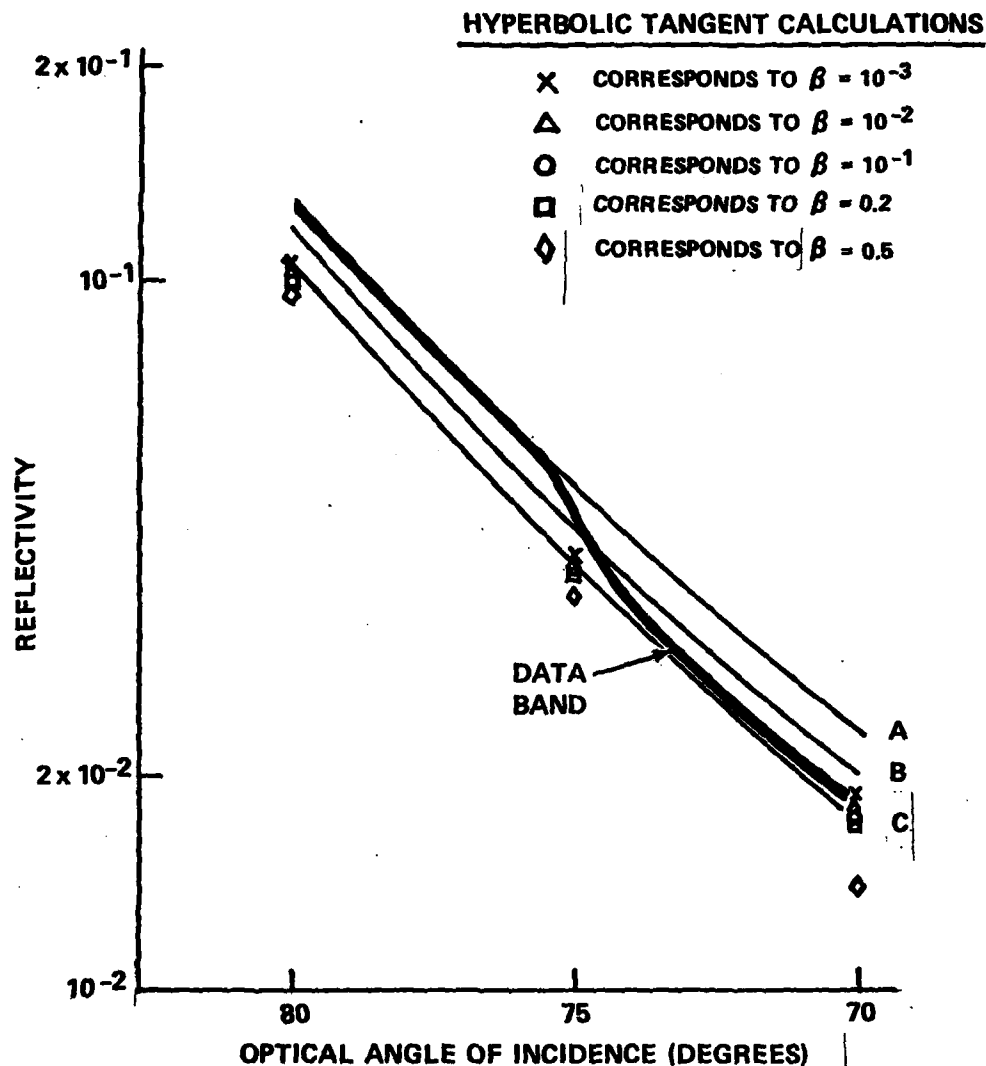
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Figure 2. Hyperbolic tangent shock front calculations for perpendicular optical polarization at 5.8 kbar. Curves A, B, and C are constant gradient model results (as in Fig. 1). All hyperbolic tangent results are for $(\Delta n)_f = 5.26 \times 10^{-2}$. $\beta = Ln_o/\lambda_o$.
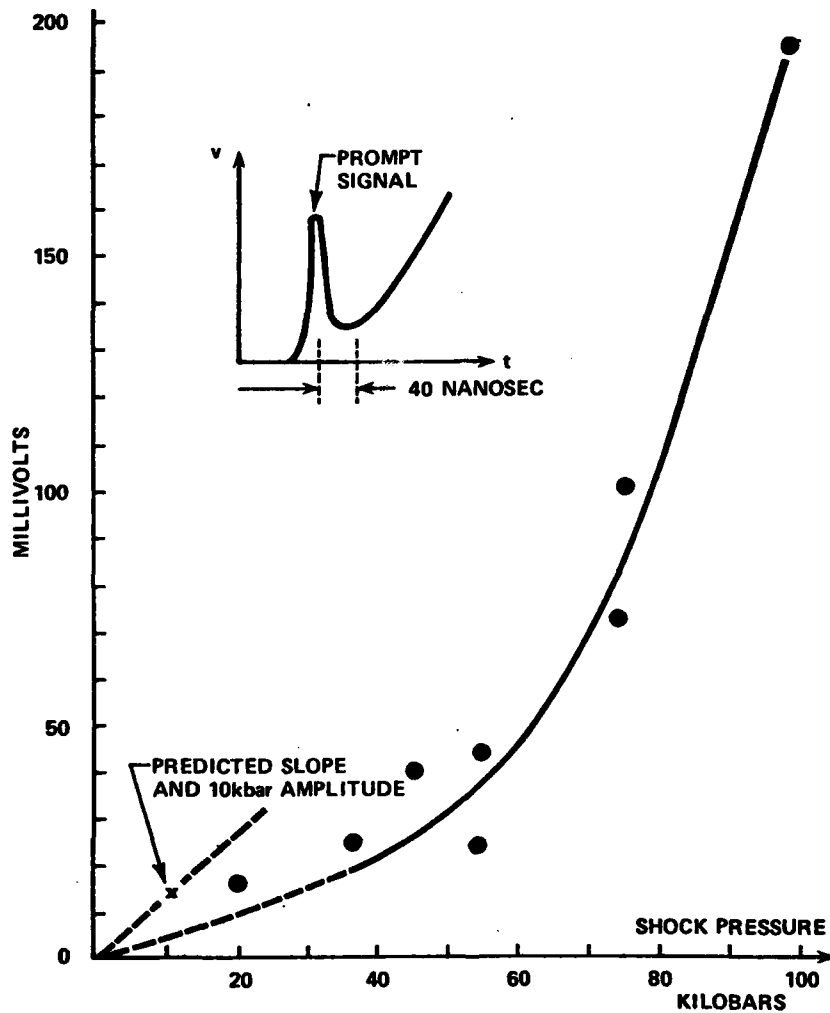
Figure 3. Averaged experimental data. The predicted slope and amplitude at 10 kbar is for net alignment of hydrogen bonded dipoles.
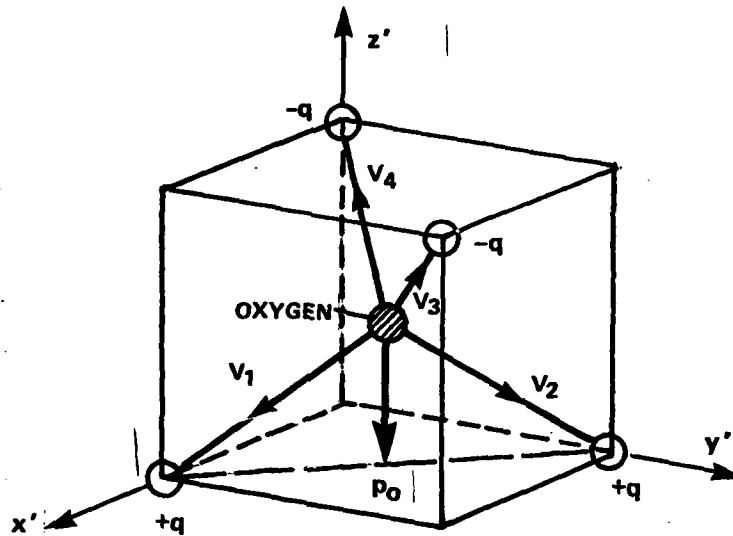
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Figure 4. The tetrahedral geometry of the rigid four-point-charge model. p_0 is the net molecular polarization, and the V_i are the coordinate vectors of the charges (and associated hydrogen masses). $q = 0.236$ electron charges.
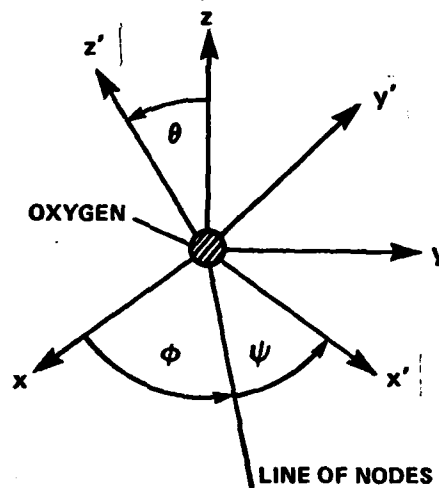

Figure 5. The Euler angle coordinates. The rigid coordinate system attached to the rigid molecule of Figure 4 is allowed to rotate within the fixed system (x, y, z). (θ, ϕ, ψ) are Euler angles, and +z is the shock propagation direction.

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REFERENCES

1. G.R. Cowan and D.F. Hornig, J. Chem. Phys. 18, 1008 (1950).

2. W.M. Flook, Jr. and D.F. Hornig, J. Chem. Phys. 23, 816 (1955).

3. P. Harris and H.-N. Presles, "The Optical Reflectivity of a Shock
    Front", Tech. Report of the Laboratoire d'Energetique et de Detonique,
    Univ. de Poitiers, 86034 Poitiers, France. Jan. 1981.

4. P. Harris and H.-N. Presles, J. Chem. Phys. 74, 6864 (1981).

5. P. Harris and H.-N. Presles, "Shock Front Rise Time in Water",
    ARRADCOM Tech. Rept. ARLCD-TR-81032, ARRADCOM, Dover, NJ 07801.
    Oct. 1981.

6. F.H. Stillinger and A. Rahman, J. Chem. Phys. 60, 1545 (1974).

7. Y. Horie, Brit. J. Appl. Phys. (J. Phys. D) 1, 1183 (1968).

8. F.E. Allison, J. Appl. Phys. 36, 2111 (1965).

9. H.A. Monterrubio, thesis "Analisis de los Resultados Experimentales
    para la determinacion del Salto de Polarizacion ΔP Inducida por
    Choque". Universidad Nacional Autonoma de Mexico (1981).

10. M.H. Rice and J.M. Walsh, J. Chem. Phys. 26, 824 (1957).

11. M. Eigen and L. De Maeyer, Proc. Roy. Soc. A 247, 505 (1958).

12. W.B. Holzappel, J. Chem. Phys. 50, 4424 (1969).

13. A. C. Mitchell, M.I. Kovel, W.J. Nellis, and R.N. Keller in High
    Pressure Science and Technology, edited by B. Vodar and Ph. Marteau
    (Pergamon, New York, 1980). Vol. 2.

14. S.D. Hamann and M. Linton, Trans. Faraday Soc. 62, 2234 (1966).

15. J.A. Pople, Proc. Roy. Soc. A 205, 163 (1951). See also D. Eisenberg
    and W. Kauzmann, The Structure and Properties of Water (Oxford Univ.
    Press, Oxford, 1969). Section 5.3.

16. D.H. Tsai and S.F. Trevino, Phys. Rev. A 24, 2743 (1981).

17. O.N. Brensov, A.N. Dremin, O.K. Rosanov, V.N. Kochnev and V.V.
    Yakushev, Soviet Electrochemistry 5, 669 (1969).

FARRIS & PRESLES

18. O.N. Brensov, A.N. Dremin, V.N. Kochnev, O.K. Rosanov, and V.V. Yakushev, Soviet Electrochemistry 7, 395 (1971).

19. V.V. Yakushev and A.N. Dremin, Soviet Phys. Chem. 45, 50 (1971).

20. V.M. Shunin, A.N. Dremin, and V.V. Yakushev, Soviet Phys. Chem. 251, 246 (1980).

21. J.J. Gilman, J. Appl. Phys. 50, 4059 (1979).

22. S.D. Hamann and M. Linton, J. Appl. Phys. 40, 913 (1969).

23. S.D. Hamann in the 1979 Novel Berzelius Symposium on "Chemistry and Geochemistry of Solutions at High Temperature and Pressure" (Pergamon, New York, 1981). Also private communications.

24. R.A. Graham, J. Phys. Chem. 83, 3048 (1979).

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